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 $\delta^{34}$ S values of selected sulfides from the Camp Smith uranium prospect and Phillips mine massive sulfide deposit and their bearing on the genesis of the deposits (New York)

By

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#### Abstract

Precambrian uraninite occurs in and around the massive sulfide body of the Phillips mine. Two different models have been proposed for the genesis of the uraninite, massive sulfide, and associated magnetite. It has been suggested that the uranium and iron oxides, as well as, the sulfides had a common magnatic source and that the deposit formed from a hydrothermal system related to that magma. A submarine volcanogenic origin for the deposit has also been suggested. This reconnaissance study of  $\delta^{34}$ S sulfide values was undertaken in an attempt to distinguish between the two models. The results which range from -4.46 to +6.71 permil show systematic variations but are insufficient to support either model; (country rocks range from +6.71 to -2.22 permil and sulfide veins, massive sulfide ores, and magnetite ores range from -4.46 to +1.28 permil).

#### Introduction

The massive stratabound sulfide deposit at Camp Smith, New York has been known since at least the mid-1800's. Development of the deposit began in the 1860's as a Cu-Ni mine, now called the Phillips mine. Shortly after operations began, plans to recover the copper and nickel were abandoned and the deposit was mined for pyrrhotite used in the production of sulfuric acid. Operations were suspended sometime in the late 1800's. Uranium was first found in the Camp Smith area in 1939 by Peter Zodac (1939). More extensive occurrences were found near the Phillips mine site in 1953 by Edward J. Chalmers. These were described by Klemic and others (1959), who concluded that the sulfides, uraninite and associated magnetite all originated from the same magmatic source and were deposited during different stages of hydrothermal activity related to that magma. Recently, Grauch (1978) suggested that the deposits are related to submarine volcanic activity and formed in a manner similar to submarine volcanogenic massive sulfide deposits. This reconnaissance study of the distribution of sulfur isotope values in and around the uranium and sulfide deposits was undertaken to evaluate (1) the existence of systematic variations that could be used to delineate a model for the formation of the deposits, and (2) the justifiability of further isotopic work.

#### Geologic setting and conceptual model for uranium concentration

The Camp Smith area is located on Precambrian rocks of the northern Hudson Highlands region of southeastern New York (fig. 1). The region has been subdivided into two distinct geologic blocks, the eastern and western highlands. The area of the Ramapo-Canopus fault system has been tentatively picked as the boundary between the two blocks. Table 1 summarizes the major Precambrian geologic events of the two blocks. The eastern highlands are

Figure 1. Index map of southeastern New York showing distribution of Precambrian rocks stippled pattern. Modified from Helenek and Mose, 1976.

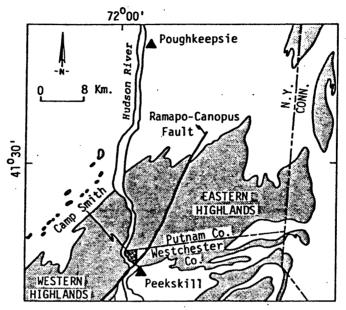


Figure 1.--Index map of southeastern New York Showing distribution of Precambrian rocks (stippled pattern). Ramapo-Canopus fault divides the Western and Eastern Highlands. Modified from Helenek and Mose (1976).

comprised predominantly of quartzofeldspathic gneisses that show the effects of several regional metamorphic events, the most intense of which was apparently upper amphibolite facies (Helenek and Mose, 1976). Lithologies in the western highlands are more varied and charnockitic gneisses, paragneisses, and granitoid plutonic rocks predominate. These rocks apparently have not been extensively affected by regional metamorphism since approximately 1100 m.y. (Helenek and Mose, 1976).

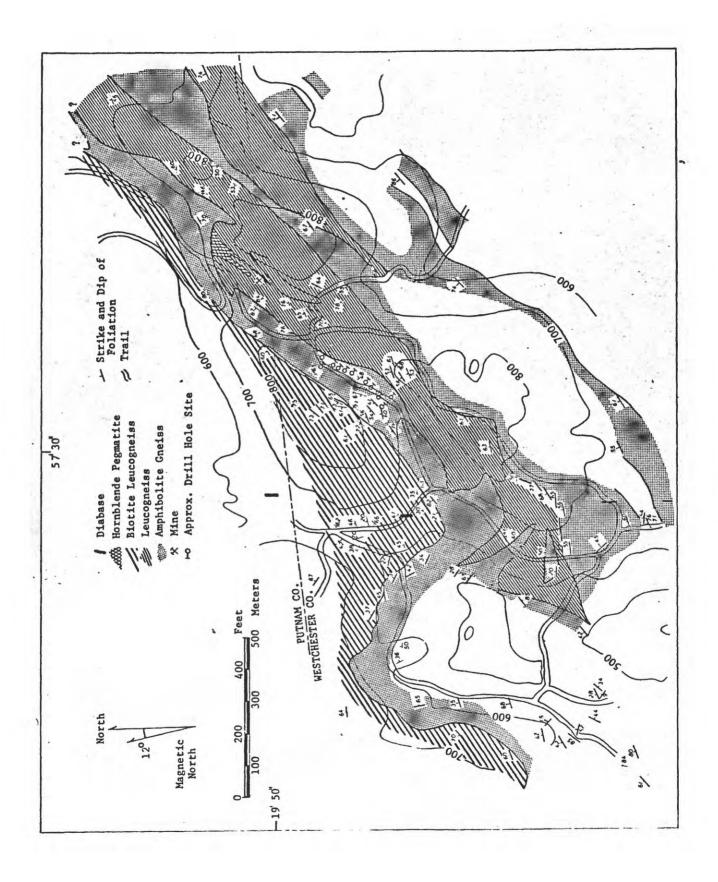
Because the lithologies present in the Camp Smith area (fig. 2) are more closely related to typical lithologies of the western highlands than they are to those of the eastern highlands, the area is considered to be a portion of the western highlands block. However, it is not clear to what extent the area was affected by the post-1100 m.y. thermal events that are recorded in the eastern highlands block. Petrographic work currently in progress (C. Nutt and R. I. Grauch, 1979, unpub. data) indicates that the Camp Smith area rocks reached at least upper amphibolite grade and that those amphibolite grade assemblages may be retrograde assemblages from granulite grade assemblages. This interpretation is consistent with the tentative assignment of the area to the western highlands block and suggests, along with the presence of metamorphosed diabase dikes, that at least the Camp Smith portion of the western highlands was affected by one or more of the post 1100 m.y. metamorphic events recorded in the eastern highlands rocks.

### Table 1.--Selected Precambrian Geologic Events in the Hudson Highlands Region

#### [Modified from Helenek and Mose, 1976. Leaders (---) indicate no data]

Time (approx.)	Western highlands	Eastern highlands
Pre-1300 m.y.		Deposition and emplacement of quartzofeldspathic sediments(?) and igneous rocks.
1300 m.y.	<b></b>	Peak of first major regional metamorphism (upper amphibolite facies).
Pre-1170 m.y.	Deposition of sediments and volcanics on pre-existing crystalline basement	· · · · · · · · · · · · · · · · · · ·
1150-1100 m.y.	Metamorphism, folding, anatexis, and plutonism (granulite facies)	<del></del>
980 m.y.		Peak of regional metamorphism.
914 m.y.	Final crystallization of anatectic granite	
Late Precambrian	Emplacement of alaskites	
Late Precambrian	Emplacement of diabase dikes	
600 m.y.		Regional metamorphism. This event has been documented to the south in the Manhattan Prong.

Figure 2. Generalized geologic map of Precambrian rocks in the northeastern portion of Camp Smith, New York. (Geologic mapping by Grauch, 1975-1977, and Klemic and others (1959).



Uranium occurs as disseminated uraninite in a variety of rock types. including scapolite-pyroxene-amphibole gneiss, magnetite-rich hornblendepyroxene gneiss, the outer zone of the massive sulfide body, and hornblende pegmatite. It also occurs in trace amounts in sulfide-rich veins that cut the amphibolite gneiss unit. Magnetite and very minor amounts of ilmenite occur in varying amounts wherever uraninite occurs except in the scapolite-pyroxeneamphibole gneiss where it has not been observed. Pyrite is always coexistent with uraninite. Chalcopyrite has been observed in the massive sulfide body, the sulfide veins, the hornblende pegmatite, and the magnetite-pyroxenehornblende gneiss. Pyrrhotite seems to be limited to the massive sulfide body, sulfide veins, and hornblende pegmatite. There is a great variety of textural relationships between the phases mentioned above. No clear-cut paragenetic sequences have yet been delineated. However, some pyrite does appear to have exsolved from pyrrhotite. This lack of consistent paragenetic relationships is attributed to the recrystallization and movement of the oxides and sulfides during metamorphism. It precludes any reliable estimation of the original phase assemblages or of "equilibrium" assemblages.

The origin of the sulfides, magnetite, and uraninite is not known. Two different models (summarized in tables 2 and 3) have been proposed. Klemic and others (1959) suggested that all the mineralization is related to a common magnatic source which generated a series of hydrothermal solutions that caused periodic mineralization. An alternative model was proposed by Grauch (1978) who suggests that the sulfides formed contemporaneously with their host sediments (a complexly interlayered sequence of carbonaceous and carbonaterich sediments and a bimodal suite of silicic and mafic volcaniclastic sediments) in a nearshore submarine volcanogenic environment similar to that proposed by Riddler (1973). The iron and uranium oxides formed at the same

#### Table 2.--Outline for magmatic model

[Geologic events based on discussion by Klemic and others (1959); limited to those observable in the immediate area of Camp Smith and having applicability to the ore genesis]

- 1. Deposition of Precambrian sediments (in part calcareous).
- 2. Metamorphism (formation of hornblende gneiss) and deformation.
- 3. Intrusion by dioritic magma during waning stages of metamorphism.
- 4a. Intrusion by magmatic solutions which formed hornblende pegmatite.
- b. Crystallization of uraninite within hornblende pegmatite and adjacent host rocks.
- c. Late, possibly pneumatolytic, stage intrusion of fluids causing (1) hornblende to alter to augite and (2) the crystallization of magnetite.
- 5. Hydrothermal deposition of sulfides (pyrite, then chalcopyrite, then pyrrhotite) within and around the same general channels through which fluids of numbers 4 (above) passed.
- 6. A separate period of intrusion resulting in the formation of oligoclasequartz pegmatite (leucopegmatite of fig. 2, minimum age of 620 m.y.).

#### Table 3.--Outline for Volcanogenic Model

[Based on Grauch (1978) geologic events limited to those observable in the immediate area of Camp Smith and having applicability to the ore genesis]

- 1a. Deposition of Precambrian sediments including calcareous, pelitic, graphitic, and volcanogenic (both basic and acidic) components.
- b. Deposition of sulfides, iron oxides, and minor amounts of phosphates at or near the seawater-sediment interface. At least one of these phases was uraniferous.
- c. Intrusion of basic dikes.
- 2. Metamorphism to at least lower granulite facies; deformation; and plutonism.
- a. Local mobilization and recrystallization of sulfides.
- b. Formation of magnetite and uraninite.
- c. Formation of hornblende pegmatite as a result of metamorphism.
- Intrusion of diabase dikes.
- 4. Metamorphism (grade unknown--possibly as high as amphibolite facies) and deformation (intensity unknown). Possible local migration and recrystallization of sulfide and oxide phases.
- 5. Intrusion of leucopegmatite.

time or slightly later in a somewhat different chemical environment, also near the seawater-sediment interface. Subsequent metamorphism and deformation resulted in the present sulfide and oxide distribution and their ambiguous paragenetic textures.

#### Sulfur isotope data

Ten samples from drill core (stored in open boxes in Maryland for 16 years), six samples from the mine dump (at least 80 years of surface exposure), and five samples from outcrops were subjected to heavy-mineral separation. The sulfides were hand picked and identified by X-ray. The  $_6$ <sup>34</sup>S values (tables 4 and 5) were then determined by standard analytical techniques using the CuO oxidation technique developed by Grinenko (1962).

#### Discussion

The data presented in table 4 show that the sulfides, at least most of them, are not in isotopic equilibrium. This is clearly demonstrated by the pyrite-pyrrhotite pair of sample 3-70a. If the two phases were in isotopic equilibrium the pyrrhotite should have a lighter  $\delta^{34}$ S value than the pyrite; it does not. A similar lack of equilibrium is shown by three other mineral pairs (see table 4, temperature column, pairs labeled N.A.). Because of the non-equilibrium demonstrated by those mineral pairs it seems unlikely that any of the pairs is in isotopic equilibrium; therefore, temperatures calculated from their isotopic fractionation values are not reliable. This lack of equilibrium is consistent with the proposed mobilization and recrystallization of the sulfide phases during one or two thermal events that post-dated their original formation.

If the magmatic model as put forward by Klemic and others (1959) is correct we would expect that the  $\delta^{34}$ S values of specific phases would fall in narrow and consistent limits within the range -13.1 to +7 permil (table 6,

#### Table 4.--6345 values of sulfides from Camp Smith, N.Y.

[J. F. Whelan, analyst]

Sample Number	Mineral <sup>1</sup>	Yield	6 <sup>34</sup> S (permil)	Theoretical 4 Temp. of formation °C	General sample category	Sample Description
1-95	ру	95.0	-0.40	1	Sulfide vein	Fine-grained hornblende gneiss with radioactive sulfide vein.
3-59	py po	84.8 102.1	-0.26 -0.25	N.A.5		Hornblende pegmatite with radioactive sulfide vein.
3-70A	py po	97.6 67.7	-1.26 -0.92	N.A.		Sulfide vein approximately 8 cm thick. Hosted by hornblende pegmatite.
3-72 1/2	Ру	93.2	+0.01			Sulfide vein approximately 1 cm thick. Hosted by hornblende pegmatite.
76-671	р <b>у</b>	92.5	-4.46	***	,	Radioactive sulfide vein cutting scapolite- pyroxene-amphibole gneiss.
3-56	ру сру	83.9 107.5	<sup>2</sup> +1.28 +0.20	372		Hornblende pegmatite with radioactive sulfide vein.
76-43	ру	90.5	-1.08	•••	Magnetite-pyroxene- amphibole gneiss	Pyroxene-amphibole gneiss with layered magnetite horizons containing disseminated sulfides.
	сру	106.1	-0.73	N.A.	,	•
76-431	РУ	96.1	-0.31		•	Pyroxene-amphibole gneiss with layered magnetite horizons containing disseminated sulfides.
76-64	<b>PY</b> .	93.6	-1.00		•	Pyroxene-amphibole gneiss with layered magnetite horizons containing disseminated sulfides.
76-65	ру Сру	91.3 ?	-1.44 -1.24	N.A.		Pyroxene-amphibole gneiss with layered magnetite- uraninite-sulfide horizons.
76-59	py po	91.1 102.1	-0.42 -1.41	280	Sulfide ore	Massive sulfide ore consisting of pyrrhotite- pyrite-chalcopyrite-magnetite-amphibole- pyroxene-apatite.
76-63	ру	98.8 96.0	-0.61 -1.35	506		Massive sulfide ore consisting of pyrrhotite- pyrite-chalcopyrite-magnetite-amphibole pyroxene-apatite.
1-36	р <b>у</b>	208.3	3 <sub>+2.96</sub>	bes	Amphibolitic gneiss	Medium grained hornblende gneiss with disseminated sulfides.
1-48	P <b>y</b>	93.1	+0.76			Coarse grained hornblende gneiss with disseminated sulfides.
1-56	ру	101.5	+3.34		Hornblende pegmatite	Hornblende pegmatite with disseminated sulfides and magnetite.
7-57	ру	95.4	+0.92			Hornblende pegmatite with disseminated sulfides and magnetite.
76-70b	p <b>y</b>	85.8	+6.71		Scapolite-pyroxene- amphibole gneiss	Scapolite-pyroxene-amphibole gneiss. Radioactive.
76-1056	ру	92.6	+2.98			Scapolite-pyroxene-amphibole gneiss. Radioactive.
76-1232	ру	89.8	<b>-2.22</b>	***	Marble	Coarse grained marble with disseminated sulfides and magnetite.
76-1104	marc	92.0	+4.44		Calcsilicate gneiss	Medium grained calcsilicate gneiss with disseminated marcasite.

<sup>1</sup>py = pyrite; cpy = chalcopyrite; po = pyrrhotite; marc = marcasite.

pyrite-pyrrhotite, 
$$To_{K} = \frac{.55 \pm .04 \times 10^{3}}{\Delta^{1/2}}; \text{ and}$$

$$pyrite-chalcopyrite, 
$$To_{K} = \frac{.67 \pm 0.04 \times 10^{3}}{\Delta^{1/2}};$$
where 
$$\Delta = .3^{4}S \text{ pyrite} = \frac{3^{4}S_{1}}{\Delta^{1/2}}$$$$

<sup>5</sup>N.A., not applicable.

<sup>&</sup>lt;sup>2</sup>Very small sample; accuracy suspect.

<sup>&</sup>lt;sup>3</sup>Very dirty due to contamination during extraction; accuracy suspect.

 $<sup>^4</sup>$ Temperatures of formation of "coexisting" pairs were obtained from the following equations of Ohmoto and Rye (in press):

Table 5.--Ranges of  $\delta^{34}$ S sulfide values for different hosts.

Host	δ <sup>34</sup> S sulfide value in permil	No. of samples
Amphibolite gneiss	+2.96 to +0.76	2 .
Scapolite-pyroxene-amphibole gneiss	+6.71 to +2.98	2
Calcsilicate gneiss	+4.44	1
Hornblende pegmatite	+3.34 to +0.92	2
Marble	-2.22	1 .
Magnetite-pyroxene-amphibole gneiss	-1.44 to -0.31	6
Sulfide ore	-1.41 to -0.42	4
Sulfide vein	-4.46 to +1.28	9

# Table 6.--Hypothetical evolution of $\delta^{34}$ S sulfide values for the magmatic model (table 2).

[This table is based on the discussion presented by Ohmoto and Rye (in press) and assumes equilibrium reactions]

The following steps would have occurred during the time period represented by steps 4 and 5 of table 2:

- 1.  $\delta^{34}S_{melt} = -3$  to +3 permil The original magma would have a value within this range.
- 2.  $\delta^{34}S_{fluid} = -3$  to +7 permil A fluid derived from the magma would have a value within this range.
- 3.  $\delta^{34}S_{H_2S} = -13$  to +7 permil;  $H_2S$  associated with a fluid with a value within the above range (#2) at temperatures above 350°C would have a value within this range.
- 4.  $\delta^{34}$ Spyrite or pyrrhotite = -12.8 to +7.9 permil
- At temperatures between 400° and 600°C pyrite or pyrrhotite formed from H<sub>2</sub>S with a value within the above range (#3) would have a value within this range. At lower temperatures the values would become heavier.
- 5.  $\delta^{34}$ Schalcopyrite = -13.1 to + 7 permil
- Chalcopyrite formed under the above conditions would have a value within this range.

step 4). The values do fall within this range. Variability of the values (table 5) could be accounted for during ore formation by changing any one or more of the systems variables (for instance,  $f_{S_2}$ ,  $f_{O_2}$ , P, T, X), or it could be accounted for by a later thermal event (which Klemic and others (1959) did not recognize). Sample density and spatial control for this reconnaissance study were not sufficient to delineate the kinds of systematic variations in  $\delta^{34}$ S values that might result from these mechanisms.

If the volcanogenic model as outlined in table 7 is valid we might expect to see systematic changes of the isotopic data as functions of stratigraphic position and horizontal variations within a single unit (see particularly Ohmoto and Rye, in press). Several investigators (cf. Mauger, 1972 and Ripley and Ohmoto, 1977) have demonstrated that these variations are preserved (at least in part) during low to intermediate grade metamorphism. There are systematic variations in the  $\delta^{34}$ S values that appear to be a function of lithology. Table 5 shows that the host rocks (with the exception of marble) and the hornblende pegmatite are enriched in  $\delta^{34}$ S as compared to the sulfide ores, magnetite-pyroxene-amphibole gneiss, and sulfide veins. If it is assumed that the original isotopic distribution has been preserved the above mentioned variation is not readily explainable in the terms of this model. However, we do not know of any data for granulite facies rocks that would either support or negate that assumption and hence, we can neither negate nor support the model.

	6 <sup>34</sup> S values in permil	Comments	Sulfide-oxide* assemblage	Comments	Geologic event (see table 3)	Comments
1.a.	10 to 20 (seawater)	Seawater in the age range of 1200-900'my would have had a value somewhere within this range (Claypool and others 1972).				
b.	-7 to +3 (sulfides)	These values are based on Sangster's (1968) empirical observation that throughout time submarine massive sulfides have had 6 345 values about 17 permil less than contemperaneous seawater.	py(I) ± cpy(I) ± po(I) + Fe hydroxide ±Ti-oxide	The original mineral assemblage is unknown but this is a reasonable assumption.	Deposition of submarine sed- iments and rocks includ- ing a signi- ficant vol- caniclastic component.	<b></b>
2.	-7 to +3 (whole rock)	If the system remained closed during metamorphism the whole rock 6345 value remained constant, and, assuming the only original sulfur-bearing phases were sulfides, the whole rock value would fall within the limits of the sulfide values (1.b above). If the system was not closed it probably became enriched in 635 because the first sulfur liberated in the reactions and lost to throughgoing metamorphic fluids was probably lighter than that left behind.	py(II) + po(II) + iss + mt(I) + ilm(I)	The original sulfides must have recrystallized. Chalcopyrite may have been superceded by iss (Cabri, 1973) and pyrite. The following are incomplete representations of reactions that probably occurred po[I] — po[II] py(I) — py(II) cpy(I) ± py(I) ± po(I) py(II) + iss Fe-hydroxide — mt(I) Ti-oxide — ilm(I) U-bearing phase — uran(I).	Metamorphism.	The peak of this event produced silicate assemblages indicative of the lower granulite facies. The temperature probably was in excess of 600°C.
3.	-7 or less to +3 or greater (sulfides)	During cooling the sulfide probably underwent a series of exsolution reactions which may or may not have produced assemblages in isotopic and (or) chemical equilibrium. Regardless, the effect would have been to extend the possible range range of 63 values. Assuming that equilibrium ceased at temperatures below 300°C, then the largest increase in range would be controlled by the largest fractionation between any two of the sulfide phase at 300°C 63 py 634 pp cpy 21.35, which is therefore the largest increase or decrease in the observed range due to equilibration.	<pre>py(III) +     po(III) +     py(IV) +     po(IV) +     cpy(II) +     mt(I) +     ilm(I) +     uran(I)</pre>	Cooling of the high temperature phases in the Cu-Fe-S system has been extensively studied but the low temperature equilibrium phase relations are incompletely understood (Craig and Scott, 1974). However, at moderate temperature (300°C) cpy-py-po may be a stable assemblage. Po(III) probably exsolved pyrite on cooling. The following are incomplete representations of equilibrium reactions that may have occurred during cooling. iss >> py(III) + po(III) + cpy(II) >> py(III)	Cooling.	It is not known what minimum temperature the rocks reached before the next thermal event (#4, table 3). But they were cold enough to undergo brittle deformation that permitted the emplacement of diabase dikes.
4.	-7 or less to +3 or greater (sulfides)	While yet another thermal event could tend to further extend the possible range of 6.3 % values it could also lead to the formation of small domains of equilibrium.	?	If this thermal event did affect the area, the above comments on metamorphism and cooling would apply to this stage except that maximum temperatures were probably much less. The degree of complexity of phase evolution would be enhanced to a large degree and it would be fruitless to speculate about possible assemblages.	Metamorphism and cooling.	The extent and nature of this thermal event in the Camp Smith area has not yet been determined.

<sup>\*</sup>py=pyrite; cpy=chalcopyrite; po=pyrrhotite; iss=intermediate solid solution; mt=magnetite; ilm=ilmenite; u=uraninite

A third model is suggested by the data. There could have been two seperate sources of sulfur, a heavy source for the host rocks and a lighter source for the ore-bearing rocks. However, the similarity of the sulfide and oxide mineralogy of the various rock types indicates that this was not probable.

#### Conclusions

- 1. Systematic variations occur in the distribution of sulfur isotope values. The sulfides of the host rocks and hornblende pegmatite are enriched in  $\delta^{34}$ S as compared to those in the sulfide ores, sulfide veins, and magnetite-pyroxene-amphibolite gneiss.
- 2. These variations do not distinguish between either of the two models proposed for the formation of the deposit. Our inability to distinguish between the models is in part a function of the reconnaissance nature of the study, and in part because the original  $\delta^{34}$ S values proposed for either model would have been nearly the same (compare the values in table 6 step 4 with those in table 7 step 1b) and in either case the sulfides have been severely and similarly altered by at least one period of metamorphism.
- 3. The apparently systematic variations in the  $\delta^{34}$ S values suggest that further isotope studies, combined with silicate and iron-titanium oxide studies currently underway, may lead to an explanation for the formation of the deposit. However, the lack of outcrop in the area precludes the kind of systematic sampling program necessary to support such work. Therefore, further isotopic work must wait for systematic drilling in the area.

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